The LLNL AMS Facility

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Abstract

The AMS facility at Lawrence Livermore National Laboratory (LLNL) routinely measures the isotopes ^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{59,63}Ni, and ^{129}I. During the past two years, over 30,000 research samples have been measured. Of these samples, approximately 30% were for ^{14}C bioscience tracer studies, 45% were ^{14}C samples for archaeology and the geosciences, and the other isotopes constitute the remaining 25%. During the past two years at LLNL, a significant amount of work has gone into the development of the Projectile X-ray AMS (PXAMS) technique. PXAMS uses induced characteristic x-rays to discriminate against competing atomic isobars. PXAMS has been most fully developed for ^{63}Ni but shows promise for the measurement of several other long lived isotopes. During the past year LLNL has also conducted an ^{129}I interlaboratory comparison exercise.

Recent hardware changes at the LLNL AMS facility include the installation and testing of a new thermal emission ion source, a new multi-anode gas ionization detector for general AMS use, re-alignment of the vacuum tank of the first of the two magnets that make up the high energy spectrometer, and a new cryo-vacuum system for the AMS ion source. In addition, we have begun design studies and carried out tests for a new high-resolution injector and a new beamline for heavy element AMS.

Introduction

In late 1989, AMS operations at Lawrence Livermore National Laboratory (LLNL) began on our FN tandem accelerator[1]. At that time about 20% of the available accelerator time was devoted to AMS, with concentration on ^{14}C measurements for bioscience and geoscience applications. Since 1989, AMS operations at LLNL have increased such that AMS measurements and AMS development activities at LLNL now consume approximately 80% of available accelerator time. We now routinely measure the isotopes ^{3}H, ^{7}Be, ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{59,63}Ni, and ^{129}I and have either in-house or collaborative programs in archaeology, bioscience, environmental science, global climate change, geoscience, materials science, and non-proliferation technologies. In this paper, a summary of AMS operations and spectrometer improvements that have occurred at LLNL during the past two years is given and future plans are discussed.

AMS Operations at LLNL

The performance of the LLNL AMS spectrometer over the past two years is shown in Table 1.

Table 1
AMS Operations at LLNL 10/93-9/95

Isotope	Stable Ion Current	Research Background	Accuracy	Number of Samples
^{14}C BioSci	100 μA ^{12}C$^-$	1×10^{-14}	2%	9110
^{14}C Natural	120 μA ^{12}C$^-$	45ka	0.7%	13900
^{3}H	15 μA ^{1}H$^-$	2×10^{-15}	10%	*450
^{10}Be	4 μA ^{9}BeO$^-$	1×10^{-14}	3%	2700
^{26}Al	1 μA ^{27}Al$^-$	2×10^{-15}	4%	1050
^{36}Cl	20-40 μA ^{37}Cl$^-$	3×10^{-15}	3%	1630
^{41}Ca	500 nA ^{40}CaF$_3^-$	3×10^{-13}	5%	*500
^{59,63}Ni	5 μA ^{58}Ni$^-$	2×10^{-11}	10%	*10
^{129}I	25 μA ^{129}I$^-$	5×10^{-14}	5%	*880
				30230

*plus several development samples

LLNL has measured over 30,000 research samples during the past two years. Of these samples, approximately 30% were ^{14}C bioscience tracer samples, 45% were ^{14}C samples for archaeology and the geosciences, and the other isotopes constitute the remaining 25%. The LLNL bioscience AMS program[2] and specific geoscience projects at LLNL[3,4,5] are discussed elsewhere.

During the past two years, a significant amount of work at LLNL has gone into the development of the Projectile X-ray AMS (PXAMS) technique. PXAMS uses induced characteristic x-rays to discriminate against competing

atomic isobars. PXAMS has been most fully developed for ^{63}Ni but shows promise for the measurement of several other long lived isotopes. During the past year LLNL has also conducted an ^{129}I interlaboratory comparison exercise. Both the PXAMS work[6] and the ^{129}I Interlaboratory Comparison Exercise[7] are discussed elsewhere.

Precision and Accuracy of ^{14}C AMS Measurements

Under normal conditions, we make 3-5 measurements to 1% precision (10,000 counts) on a typical unknown sample. After normalization to the Oxalic Acid Standard (OX1), these 3-5 measurements result in ^{14}C values that are nominally precise to 0.6-0.8 per mil. To further characterize the precision and accuracy of our ^{14}C AMS measurements, we routinely measure targets from several secondary standards in each wheel of samples. Figure 1 shows results obtained from New Oxalic Acid Standard (OX2) targets that were graphitized and measured at LLNL from March, 1995 to April, 1996.

Under our present measurement scheme, ^{14}C/^{13}C ratios of OX2 secondary standards are measured about 8 times during the course of a typical run. Each measurement typically takes 20 seconds and is done is to 1% counting statistics. After normalization to the average of four OX1 ^{14}C/^{13}C ratios, background and fractionation corrections are applied, and Fraction Modern values are calculated. Error bars shown in Figure 1 are based on OX2 and OX1 counting statistics uncertainties with an uncertainty contribution from the background correction. (Note that the uncertainty value we normally quote for an unknown is the larger of such a counting-statistics based estimate and an estimate based on the scatter of the individual determinations of the ^{14}C/^{13}C ratio of a target. Hence, the error bars shown in Figure 1 are, in some cases, smaller than we would normally quote.).

The precision of the value obtained for a single target can be estimated from the scatter of the 83 OX2 values in this data set. This estimate of ± 0.0064 (1 Standard Deviation) indicates that the measurement of a single target is precise to ± 4.8 per mil. Comparison of this estimate to the counting statistics based estimate of the typical uncertainty in the measurement of a single target of ± 0.0055 (4.1 per mil) indicates that counting statistics uncertainties account for all but approximately 2.5 per mil of the observed scatter in the OX2 data set.

The accuracy of the value obtained for a single target can be estimated by comparing the per cent Modern Carbon (pMC) value derived from the average of 83 OX2 values of 133.9 ± 0.6 (1 Standard Deviation) to the accepted OX2/OX1 value of 134.1[8]. The agreement between these pMC values indicates that any systematic offset in our measurements is less than 1.5 parts per thousand.
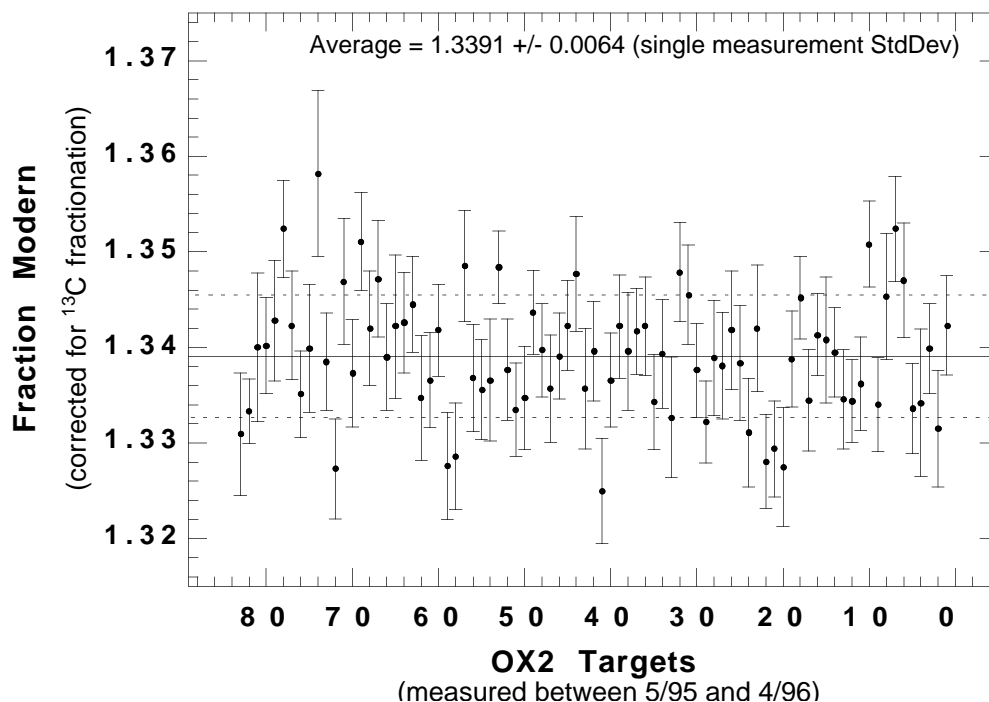
Figure 1. Fraction Modern values obtained from 83 OX2 targets at LLNL from March, 1995 to April 1996. Each data point represents a single OX2 target. Error bars are based on counting statistics uncertainties. Data are arranged in chronological order left to right. The solid line indicates the average of the data while the dashed lines indicate the ±1 Standard Deviation uncertainty calculated from the data for a measurement on a single target.

Spectrometer Improvements

One of the recent hardware changes to our facility was the installation of a new final detector housing shown in Figure 2. While the heart of this new detector housing is a fairly standard five anode gas ionization detector, other parts of the detector housing are unique in that they were specifically designed for ease of use and maintainability. For instance, pumped volume of the detector housing was minimized to decrease pump down time, access to the entrance window of the gas ionization detector is through a quick-entry port so that we can change detector windows in a matter of minutes, dedicated ports were provided for solid-state, time-of-flight, and X-ray detectors, and special attention was paid to electronic noise shielding.

During the past year, we found it necessary to realign the vacuum tank of the first of the two magnets that make up the high energy spectrometer. For some time, it had been observed during ^{10}Be AMS measurements that not all of the ^{9}Be beam was being transmitted into the off-axis stable isotope Faraday cup. Transmission efficiency varied slightly from sample-to-sample and day-to-day, with a long-term trend (1-2 year) towards increasing ^{9}Be loss. Upon detailed examination of the problem (including the running of several

beam transport models), it was determined that the ^{9}Be beam was scraping the side of the vacuum tank of the first high energy spectrometer magnet. Sample-to-sample variations and small differences in tuning were causing the varying transmission, but the cause of the long-term trend is still not understood. To resolve the problem, we partially disassembled the first spectrometer magnet and off-set the vacuum tank approximately 2.0 cm to the low mass side. This resolved the varying ^{9}Be transmission problem and we now have 100% ^{9}Be transmission into the off-axis Faraday cup.
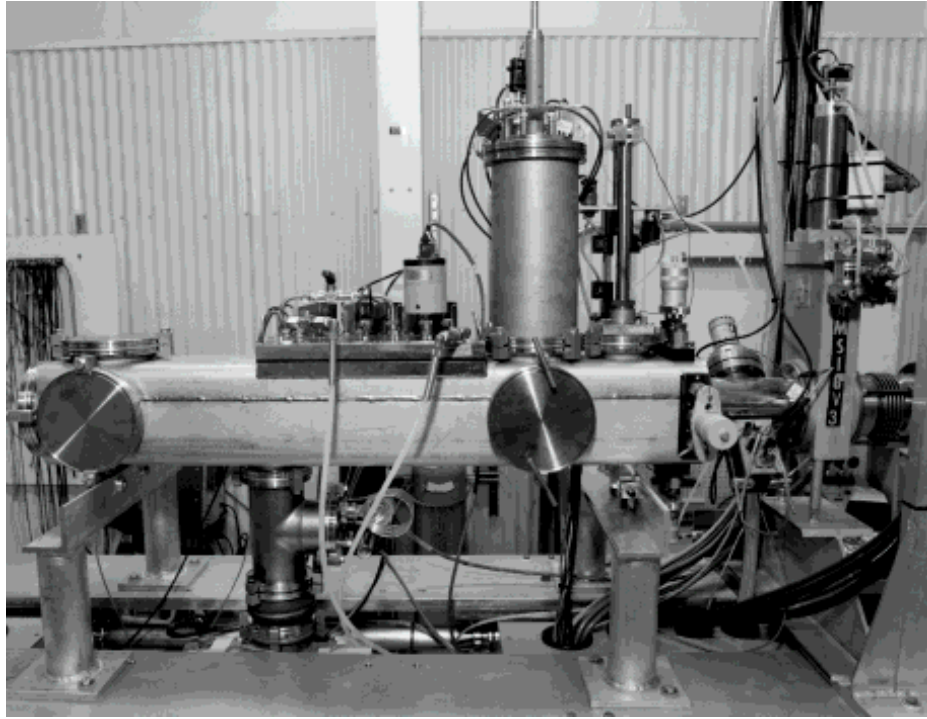

Figure 2. The LLNL AMS detector housing.

Another change to the LLNL AMS facility was the installation of a CTI-Cryogenics On-Board 8 Cryopump on the AMS ion source. This cryopump replaced the turbomolecular pump used previously. While the new cryo-system was installed primarily to eliminate the possibility of turbomolecular pump oil back-streaming and contaminating small ^{14}C samples, the cryo-pump has had several other advantages. The base pressure in the ion source has been reduced from $1x10^{-6}$ Torr to about $3x10^{-7}$ Torr, pump down time after a sample wheel change has been cut nearly in half, and because of the excellent water vapor pumping capability of a cryo-pump, the contribution of ^{12}CH$^-$ beam to mass 13 beam (as measured on our low energy Faraday cup) has been reduced approximately 50%. It also appears that the cryo-system has substantially reduced ^{36}S backgrounds in ^{36}Cl AMS measurements. The cryo-system is fully interlocked and to date we have had no major vacuum accidents and we have not experienced any problems with

cesium vapor or sample or source outgassing loads.

Thermal Emission Ion Source

During the past two years we have also installed and tested a thermal emission ion source on our accelerator. This ion source was installed as part of a joint Pacific Northwest National Laboratory/LLNL project that proposed to combine the high sample-efficiency of a thermal emission ion source with the high isotopic ratio sensitivity of AMS. With the combination of thermal emission ion source and AMS, we had hoped to demonstrate that isotopic ratios of $^{129}I/^{127}I$ as low as 10^{-11} could be measured from samples as small as a few μg.

The ion source for the project was a commercial, 20 sample, thermal emission ion source purchased from VG Isotech. The ion source was installed on a high voltage platform on a new beamline off of our 90° AMS injection magnet. The beamline utilized a National Electrostatics Corporation electrostatic quadrupole triplet lens to match the 'line shaped' beam emittance of the thermal emission ion source to the 'circular shaped' beam acceptance our accelerator. A picture of the thermal emission ion source and injection line is shown in Figure 3.
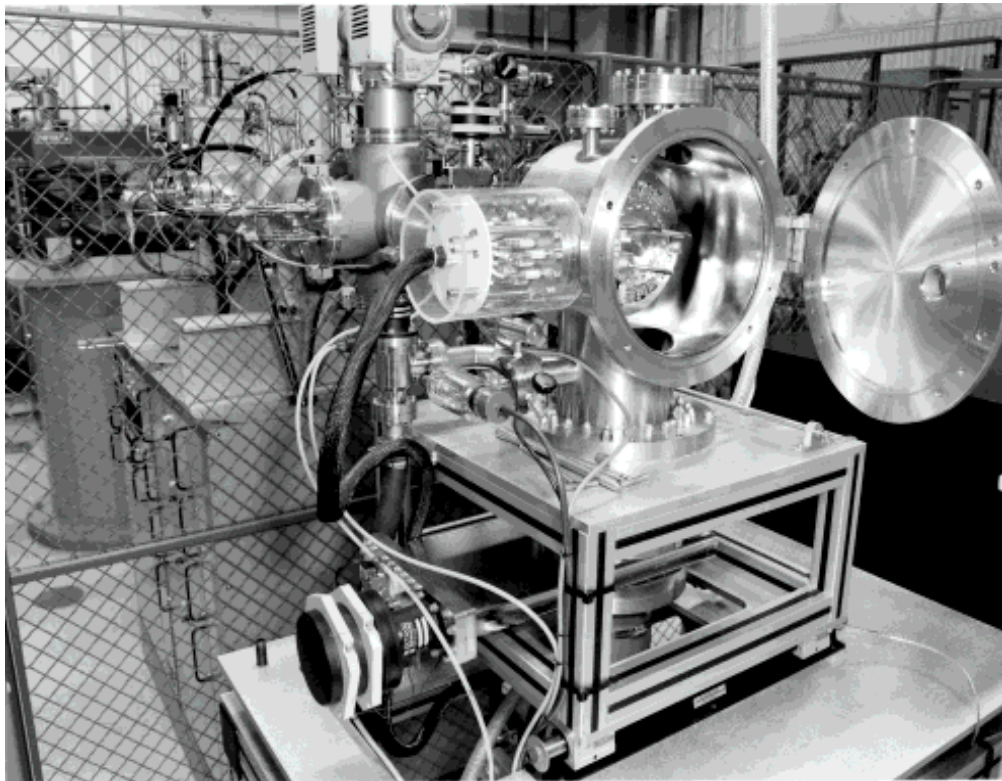

Figure 3. The thermal emission ion source and injection line.

Unfortunately, project results were disappointing. Using the thermal emission ion source, we were never able to obtain more 1 or 2 nA of $^{127}I^-$ current. With such small ion currents, it was almost impossible to 'tune' or set machine parameters using our standard Faraday cups and beam profile monitors. In the end, we were never able to obtain $^{129}I/^{127}I$ ratios using the thermal emission ion source in conjunction with our AMS spectrometer and the project was canceled.

Future Plans

Currently, we are building a new high-resolution high-mass AMS injector and beamline. This injector and beamline will be used to develop AMS for those long lived isotopes with masses between 79 and 244. The injector part of this system will be built first and will utilize a Danfysik electrostatic analyzer (Model 263, 90° deflection angle, 750 mm bend radius, 100 mm plate separation, mean beam rigidity of 75 kV), and an ion source[9]/sample changer[10] identical to our present setup. This new injector should be operational sometime in late 1996. The high-mass ion detection part of this system is currently being designed. Initially, this beamline will use a 30° magnetic analysis and a Wien or velocity filter to reduce count rates from various charge-exchanged ions. As funding permits, however, we will replace the Wien filter with a large Danfysik electrostatic analyzer (Model 264, 30° deflection angle, 4375 mm bend radius, 50 mm plate separation, mean beam rigidity of 7 MV).

Other developments in our laboratory will help to increase AMS throughput by reducing the current FN beam time bottleneck. Currently, we are installing a new stand-alone microprobe system in our facility. This system will utilize a 1.7 MV National Electrostatic Corporation 5SDH-2 tandem accelerator and will be used for micron-scale materials analysis studies. Currently, the microprobe group uses approximately five FN accelerator beam days per month and when the new microprobe facility comes 'on-line' in the fall of 1996, microprobe activities on the FN accelerator will decrease significantly. We have also solicited, and are awaiting a funding decision, on a new dedicated ^{14}C bioscience AMS machine. This machine will be developed in conjunction with High Voltage Engineering Europa and will utilize a 750 kV tandem accelerator. Together these developments will free up another 7 to 9 days of FN beam time a month allowing significantly more time for AMS 'production' or development activities.

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